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# NONDESTRUCTIVE EVALUATION OF NUCLEAR-GRADE GRAPHITE

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**ABSTRACT.** The material of choice for the core of the high-temperature gas-cooled reactors being developed by the U.S. Department of Energy's Next Generation Nuclear Plant Program is graphite. Graphite is a composite material whose properties are highly dependent on the base material and manufacturing methods. In addition to the material variations intrinsic to the manufacturing process, graphite will also undergo changes in material properties resulting from radiation damage and possible oxidation within the reactor. Idaho National Laboratory is presently evaluating the viability of conventional nondestructive evaluation techniques to characterize the material variations inherent to manufacturing and in-service degradation. Approaches of interest include x-ray radiography, eddy currents, and ultrasonics.

**Keywords:** Graphite, Ultrasonic, Eddy Current, X-ray Radiography

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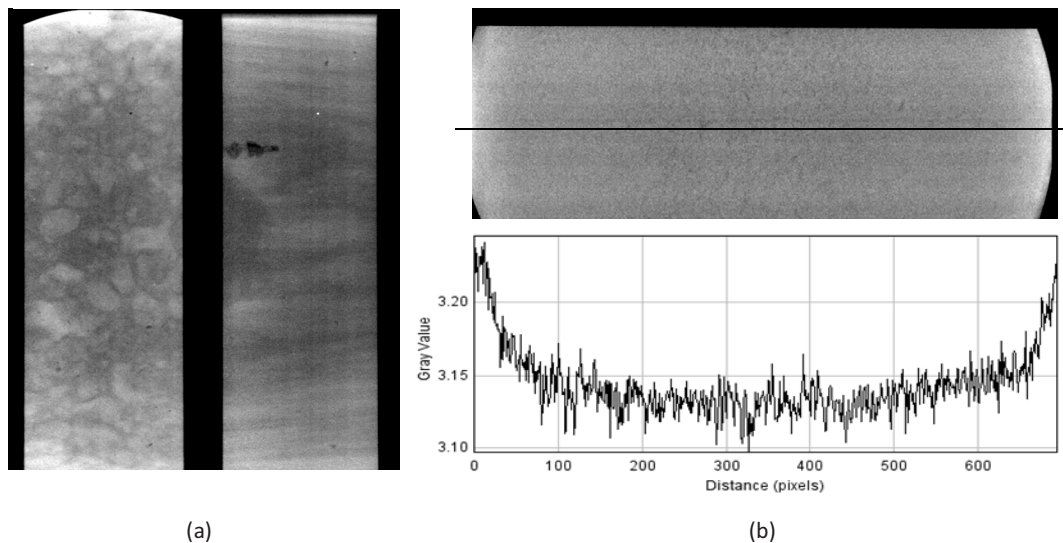
## INTRODUCTION

Nuclear-grade graphite is a composite material made from petroleum or a coal-tar-based coke and a pitch binder. Manufacturing graphite is an iterative process of baking and pitch impregnation of a formed green billet prior to final graphitization, which occurs at temperatures greater than 2500°C. The impregnation and rebake step is repeated several times until the desired product density is obtained. Manufacturing times for a lot of graphite can range from 6–9 months [1]. Integral to this process is the use of isotropic cokes and a forming process (i.e., isostatically molded, vibrationally molded, or extruded) that is intended to obtain an isotropic or near isotropic material. However, the source, size, and blend of the starting materials—as well as the forming process of the green billet—will impart variations within the final product. There will be density variations from the billet surface inward and different physical properties with and transverse the forming direction. Material variations are expected within individual billets as well as billet-to-billet and lot-to-lot. In addition to the material variation inherent to the manufacturing process, graphite will experience changes in volume, mechanical strength, and thermal properties while in service in a nuclear reactor. It is therefore recognized by the U.S. Department of Energy (DOE)'s Next Generation Nuclear Plant (NGNP) Program that nondestructive evaluation (NDE) technologies will need to be utilized to characterize the graphite in the as-fabricated through the in-service condition as a means to ensure material properties and in-service integrity.

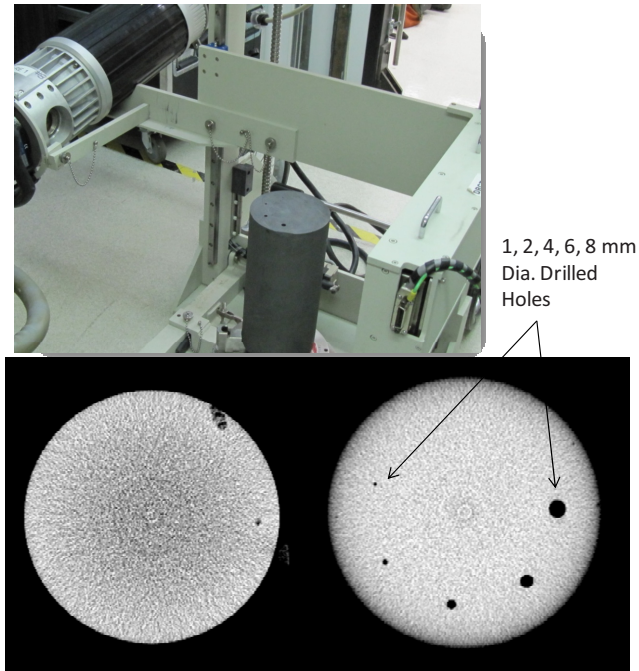
Although graphite has been used in nuclear reactors for over 60 years, only a limited amount of work has been performed to develop NDE technologies for inspecting graphite [2–4]. There are few ASTM/ASME standards or accepted measurement techniques specifically directed towards graphite. In fact, the nuclear-grade graphite used before in the U.S., H-451, is no longer made and new graphite types are being qualified via the NGNP program. For these reasons, Idaho National Laboratory (INL) is initially evaluating conventional NDE technologies to understand and or demonstrate their viability for inspecting graphite materials prior to investigating potentially viable but undeveloped technologies. The technologies being evaluated include x-ray radiography, eddy currents, and ultrasonics. Note that the specifics of the type and size of material anomalies and/or defects that will be detrimental to service have not yet been fully established.

### **X-Ray Radiography/Tomography**

X-ray radiography is well-suited to characterize the distribution of porosity in graphite components, as well as the detection of high density inclusions, large pores, cracks, or physical defects produced during the machining of core components. The candidate reactor graphite types now being evaluated by NGNP range in density from 1,770–1,870 kg/m<sup>3</sup> [5], thereby permitting the use of industrial based x-ray imaging systems for moderate size components. However, large billets may require high energy sources and detectors for adequate x-ray penetration and imaging. Although 2D projection radiography will be adequate for simple geometries (see Figure 1), computed tomography (CT) will be required to fully address billets having surface irregularities and finished components with complex structures.



**FIGURE 1.** (a) Orthogonal radiographs of a 165 mm wide x 158 mm thick graphite sample cut from a 483 diameter extruded billet. Although the porosity is excessive in nature, the sample does provide an example of the ability of x-ray radiography to image porosity and its unique character produced by the extrusion process. Note that the type and grade of this graphite is unknown and is not typical of current reactor-grade material. (b) A radiograph taken parallel to the extrusion axis of a 165 mm wide x 158 mm thick reactor-grade graphite sample cut from a 470 mm diameter billet. Note the variation in density across the diameter of the billet.

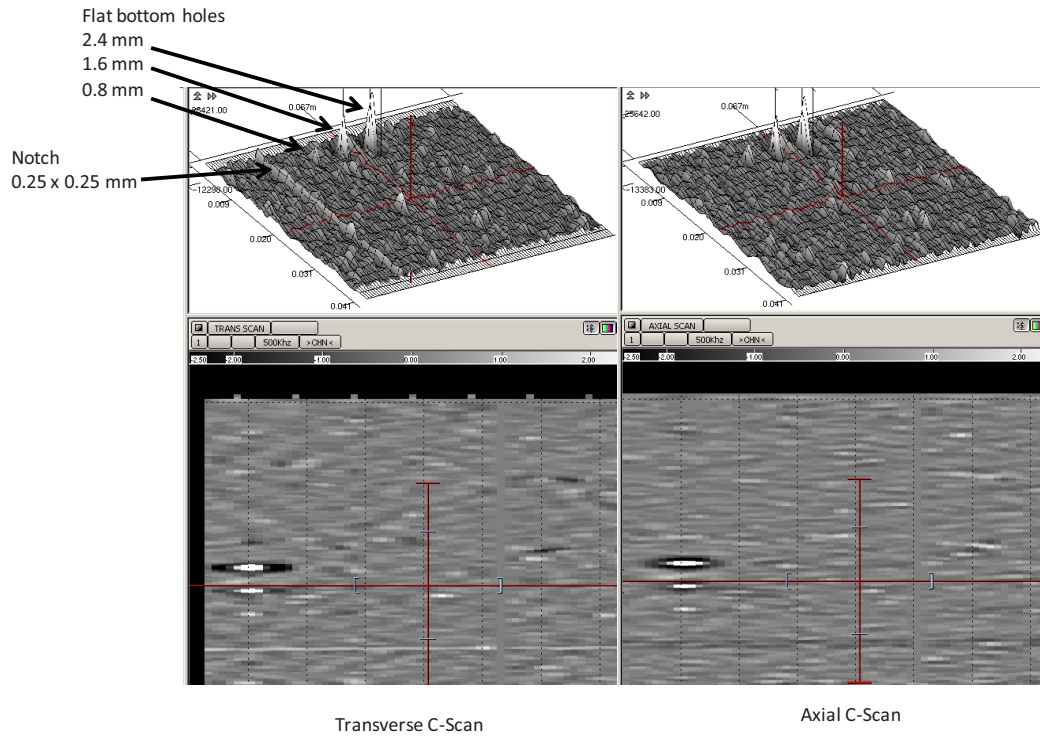


**FIGURE 2.** Horizontal x-ray CT slices of a 152 mm diameter graphite test sample. The CT slices are located at two different vertical locations: below and through the vertically drilled holes.

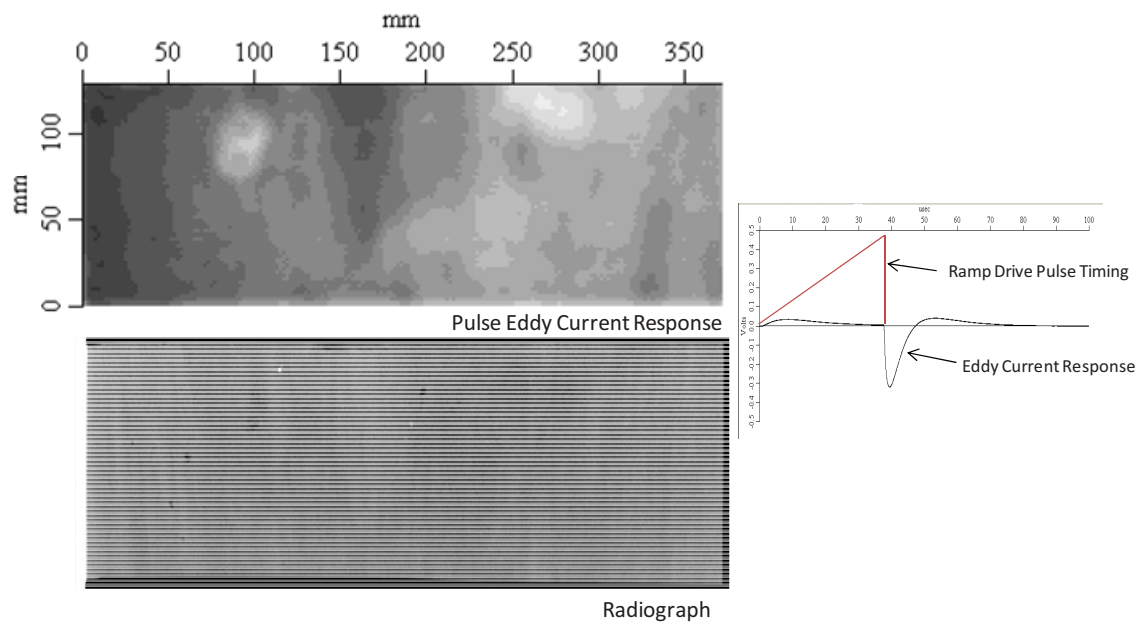
Figure 2 provides an example of the 3D information that can be obtained from computed tomography using a 153 mm diameter extruded graphite test sample. Observed in the CT slices taken from different heights along the cylinder are natural defects as well as machined holes ranging in diameter from 1 mm to 8 mm. The x-ray projections used to construct the slices were obtained using a 1.5 mm focal spot size source running at 220 kV and 3.0 mA. The detector was a linear array with a 0.45 mm element pitch, the source to detector distance was 932 mm, and the source to the center of the rotation distance was 621 mm, thereby yielding a magnification of approximately 1.5X.

## **Eddy Currents**

NGNP graphite candidates have electrical conductivities that range from  $9.8 \times 10^4$  to  $1.3 \times 10^5 \text{ Sm}^{-1}$  [5]. This makes eddy currents a viable approach to perform surface/near surface inspections for cracking and/or material damage due to radiation, mechanical loading, or oxidation. Figure 3 provides a C-scan of a medium-grain vibrationally molded graphite sample containing flat bottom holes and a full width notch. The data was collected using a 64 element driver-pickup array probe being driven at 500 kHz. The coil elements are arranged and used in a manner to collect two orthogonal C-scans that have sensitivities in the axial and transverse direction to the scan axis. Due to the test frequency and coil element size (i.e., 2 mm coils / 2.5 mm pitch) little information is gained relevant to subsurface material conditions. Although limited by test frequency skin depth and the ability to physically project a magnetic field from an induction coil, eddy currents can also be used to obtain near surface information. Figure 4 shows a C-scan of a 51 mm thick graphite plate containing surface breaking defects and variable porosity. A 25 mm diameter probe driven by a ramp pulse was used to collect the data. The C-scan image provided is the peak amplitude of the eddy current response.



**FIGURE 3.** Eddy current surface inspection of a graphite sample containing flat bottom holes (i.e., 2.4, 1.6, and 0.8 mm dia., respectively) and a full width notch (i.e., 0.25 mm deep  $\times$  0.25 mm wide). A 64-element driver-pickup array probe was used to simultaneously collect the two C-scan images.



**FIGURE 4.** Pulsed eddy current inspection of a graphite plate containing variable porosity. Due to the increased size of the probe (25 mm), and the 70 V peak amplitude drive signal, sufficient penetration is obtained to map the porosity distribution internal to the plate.

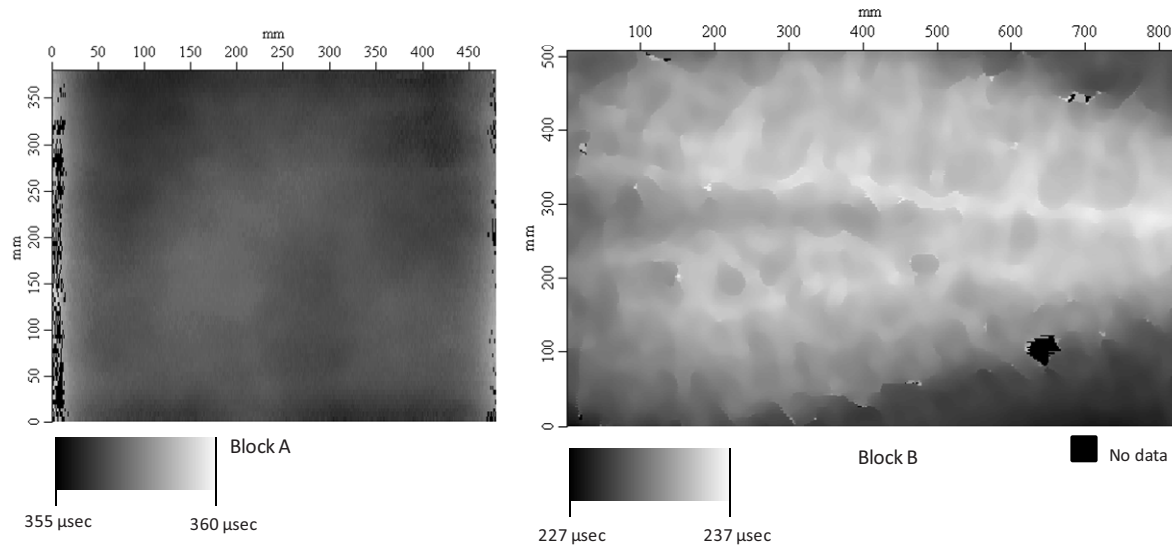
A good correlation is observed between the eddy current image and the density variations observed in the radiograph. However, note that large surface-breaking defects on the back side of the plate were not imaged, indicating that for this test configuration, magnetic field penetration was insufficient to characterize the full thickness of the plate. The primary use of eddy current testing is the evaluation of surfaces accessible by a probe.

## **Ultrasonics**

Due to the presence of porosity and its composite microstructure, graphite attenuates higher ultrasonic test frequencies. This combined with the range of acoustic velocities measured for NGNP candidate graphites ( $C_L = 2.5\text{--}2.9 \text{ kms}^{-1}$  and  $C_S = 1.5\text{--}1.7 \text{ kms}^{-1}$  [5]) result in relatively long acoustic inspection wavelengths that limit the ability of ultrasonic inspections to detect small isolated flaws. Note that since a large population of “flaws” already exist within the microstructure in the form of porosity, there is no practical need to detect small individual flaws less than or equal in size to the inherent porosity. The long acoustic wavelengths that can propagate through significant thicknesses of graphite (e.g., 2.9 mm at 1 MHz) will still be capable of detecting macro-cracks, large voids, and inclusions.

Of interest is the potential for ultrasonics to characterize the uniformity of the microstructure and detect the presence of distributed flaw structures internal to thick sections. Radiography will not have the spatial resolution to detect distributed flaws, such as micro-cracking, and eddy currents are limited to surface/near surface examinations. Changes in material microstructure or flaw structures that alter the elastic properties of the graphite will result in changes in acoustic velocity and changes in the scatter and absorption suffered by the propagating wave. Round trip time-of-flight C-scans for the back wall reflection for two different graphite blocks are presented in Figure 5. The data was collected using a 25.4 mm, 2.25 MHz contact transducer with water coupling. Both blocks had machined, parallel surfaces. Block A is a medium-grain, vibrationally molded, 464 mm thick billet with a maximum variation in velocity of  $0.03 \text{ kms}^{-1}$ . Block B is a fine-grain, isostatically molded, 305 mm thick billet with a maximum variation in velocity of  $0.11 \text{ kms}^{-1}$ . The data collected indicates that the microstructure and/or flaw population in Block B is highly variable compared to Block A. It also suggests that the mechanical properties of Block B will demonstrate a similar variability to that recorded for the acoustic velocity. Note that Block B is an experimental grade of graphite not fully developed for production.

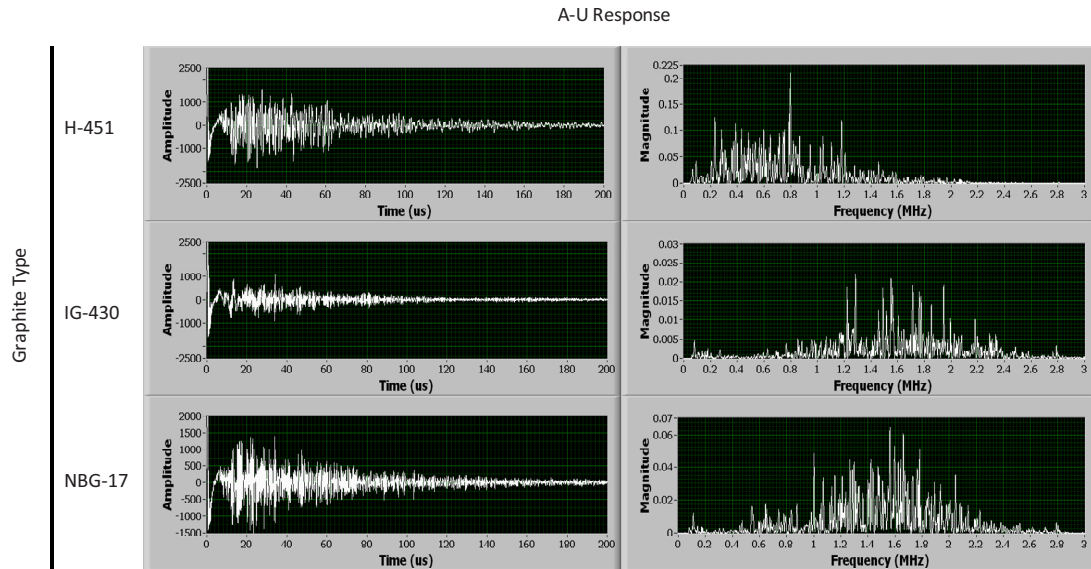




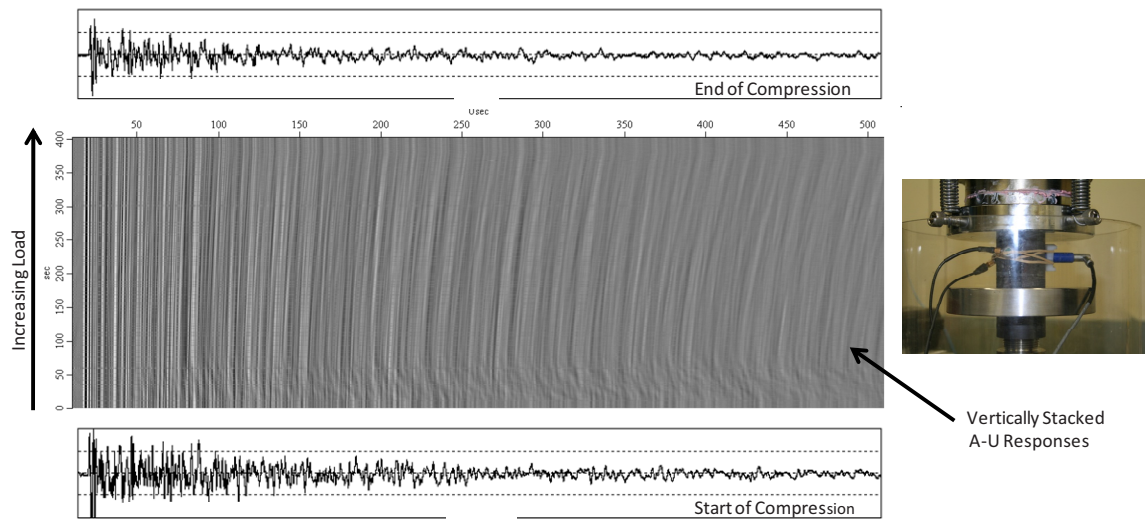
**FIGURE 5.** Back wall time-of-flight C-scan images of two different machined graphite blocks. Block B has significant variations in acoustic velocity suggesting that there will be similar variations in mechanical properties.

Acoustic wave scatter measurements also have the potential to detect variations in microstructure, as well as the presence of distributed flaw structures. Acousto-ultrasonics (A-U) is a technique initially developed in the 1970s to evaluate materials that are difficult to inspect using conventional ultrasonic techniques [6]. The approach is based on a transmit/receive concept in which the receiving transducer does not directly sense the acoustic wave introduced by the transmitter but collects the scattered energy generated as the transmit wave propagates through the local microstructure. The scatter recorded will contain characteristics specific to the microstructure and any defects structure present. This is illustrated in Figure 6 in which A-U responses collected from three different graphite grades are presented. The grain size and inherent porosity is different for each grade resulting in different frequency content and attenuation in the A-U responses. The A-U waveforms were collected from one side using 6.3 mm, 2.25 MHz contact transducers separated 20 mm. Water was used as the couplant.

Defect structures will also result in changes in the scatter recorded. As a means to better understand what will change in the recorded A-U response, a graphite sample was instrumented with transducers and compressed in a mechanical testing machine to induce internal damage. Figure 7 contains the A-U responses recorded in real-time as a sample of H-451 graphite is slowly compressed to a maximum load of 51 MPa. A 5% decrease in acoustic velocity was measured for the test sample after compression indicating that internal damage was produced. Examination of the A-U responses indicates that a rapid change in the recorded scatter occurs early in time, followed by a more gradual drift in signal timing late in the waveform as the load continues to increase. It is hypothesized that the initial flurry of activity results from the collapse of small pores. The subsequent gradual change is believed to be due to increased micro-cracking in the graphite. These results suggest that information late in the A-U waveform will need to be examined to identify the presence of micro-cracking.



**FIGURE 6.** A-U comparison of three graphite types.



**FIGURE 7.** Graphite A-U responses during compression. A 5% decrease in acoustic velocity was measured after compression testing indicating that internal damage was produced in the test sample.

## CONCLUSIONS

It has been demonstrated that the application of conventional NDE technologies provide information relevant to the condition of nuclear-grade graphite. However, additional research will be required to understand and quantify the results obtained and implement the measurements external to INL. At this time a combination of NDE techniques will be required to evaluate bulk graphite and manufactured components during construction and in-service inspection.



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